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(54) **R-T-B sintered permanent magnet**

(57) An R-T-B sintered permanent magnet having a composition including 28-33 weight % of R, and 0.5-2 weight % of B, the balance being substantially T and inevitable impurities, wherein R is at least one rare earth element including Y, at least one heavy rare earth element selected from the group consisting of Dy, Tb and Ho being indispensable, and T is Fe or Fe and Co, the permanent magnet having a crystal structure comprising first  $R_2T_{14}B$ -type, main-phase crystal grain particles having a higher heavy rare earth element concentration than that of a crystal grain boundary phase, and second  $R_2T_{14}B$ -type, main-phase crystal grain particles having a lower heavy rare earth element concentration than that of the crystal grain boundary phase.

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of nitrogen and 0.3 weight % or less of Ca as inevitable impurities.

[0013] The R-T-B sintered permanent magnet of the present invention is produced, for instance, by the steps of mixing of two types or more of alloy powder having substantially the same composition except for the difference in a ratio of heavy rare earth elements (Dy, etc.) / light rare earth elements (Nd, Pr, etc.) with the same total amount of the rare earth elements, molding in a magnetic field, sintering, heat treatment, and if necessary, machining, finish working such as barreling, etc., and surface treatment such as Ni plating etc. Depending on the compositions of the above two types or more of alloy powder and the final composition of the R-T-B sintered permanent magnet, the optimum sintering conditions are selected to strictly control the diffusion of heavy rare earth elements such as Dy in the crystal structure of the sintered magnet. As a result, the crystal structure has a characteristic concentration distribution of heavy rare earth elements such as Dy in the  $R_2T_{14}B$ -type, main-phase crystal grain particles (substantially in center portions) and the crystal grain boundary phase, containing  $R_2T_{14}B$ -type, main-phase crystal grain particles having a higher concentration of heavy rare earth elements such as Dy than that of the crystal grain boundary phase, and  $R_2T_{14}B$ -type, main-phase crystal grain particles having a lower concentration of heavy rare earth elements such as Dy than that of the crystal grain boundary phase.

[0014] The R-T-B sintered permanent magnet having such a sintered crystal structure has extremely larger  $B_r$  and  $(BH)_{max}$  than those of the R-T-B sintered permanent magnet produced by the single method, though its coercivity  $iH_c$  is slightly smaller than that of the latter. Though the correlation between such high magnetic properties and the concentration distribution of heavy rare earth elements such as Dy has not been fully clarified yet, it is presumed that the  $R_2T_{14}B$ -type, main-phase crystal grain particles having a higher concentration of heavy rare earth elements such as Dy than that of the crystal grain boundary phase contributes to achieving high  $B_r$ , while the  $R_2T_{14}B$ -type, main-phase crystal grain particles having a lower concentration of heavy rare earth elements such as Dy than that of the crystal grain boundary phase contributes to achieving high  $iH_c$  close to that obtained by the single method.

#### BRIEF DESCRIPTION OF THE DRAWINGS

##### [0015]

Fig. 1 is a graph showing the relation between a sintering temperature and magnetic properties ( $B_r$ ,  $iH_c$ ) in the R-T-B sintered permanent magnets of EXAMPLE 1 and COMPARATIVE EXAMPLE 1;

Fig. 2 is a graph showing the relation between a sintering temperature and magnetic properties ( $B_r$ ,  $iH_c$ ) in the R-T-B sintered permanent magnets of EXAMPLE 2 and COMPARATIVE EXAMPLE 2;

Fig. 3 is a graph showing the relation between a sintering temperature and magnetic properties ( $B_r$ ,  $iH_c$ ) in the R-T-B sintered permanent magnets of EXAMPLE 3 and COMPARATIVE EXAMPLES 3 and 4;

Fig. 4 (a) is a schematic view showing the crystal structure of the R-T-B sintered permanent magnet of EXAMPLE 7;

Fig. 4 (b) is an EPMA photograph showing the concentration distribution of Dy in the crystal structure of the R-T-B sintered permanent magnet of EXAMPLE 7;

Fig. 4 (c) is an EPMA photograph showing the concentration distribution of Nd in the crystal structure of the R-T-B sintered permanent magnet of EXAMPLE 7;

Fig. 4 (d) is an EPMA photograph showing the concentration distribution of Pr in the crystal structure of the R-T-B sintered permanent magnet of EXAMPLE 7;

Fig. 5 is a graph showing the particle size distribution of main-phase crystal grain particles in the R-T-B sintered permanent magnet of EXAMPLE 7;

Fig. 6 is a schematic view showing the crystal structure of the R-T-B sintered permanent magnet of COMPARATIVE EXAMPLE 5; and

Fig. 7 is a graph showing the particle size distribution of main-phase crystal grain particles in the R-T-B sintered permanent magnet of COMPARATIVE EXAMPLE 5.

#### DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

##### [1] R-T-B sintered permanent magnet

###### (A) Composition

###### (a) Main components

[0016] The R-T-B sintered permanent magnet of the present invention comprises main components comprising 28-33 weight % of R, 0.5-2 weight % of B and the balance being substantially T, and inevitable impurities. In addition, 0.01-

tically like Al and Ga. On the other hand, when the content of Cu is less than 0.01 weight %, sufficiently effects of improving iHc and corrosion resistance cannot be obtained.

[0026] As described above, the content of the  $M_2$  element, either Al, Ga or Cu, is 0.01-0.3 weight %.

## 5 (b) Inevitable impurities

[0027] The inevitable impurities include oxygen, carbon, nitrogen, calcium, etc. When two types or more of R-T-B alloys having different heavy rare earth element contents are produced by a reduction diffusion method, Ca enters into them as an inevitable impurity. The reduction diffusion method is a method for producing the alloy powder by reducing  
10 powder of rare earth element oxides with a reducing agent (Ca), and then subjecting the resultant rare earth element metal powder to mutual diffusion with other main component metals.

[0028] The content of oxygen is preferably 0.6 weight % or less, the content of carbon is preferably 0.15 weight % or less, the content of nitrogen is preferably 0.15 weight % or less, and the content of calcium is preferably 0.3 weight % or less. When the content of each inevitable impurity exceeds each above upper limit, the R-T-B sintered permanent  
15 magnet has decreased magnetic properties. More preferable contents of inevitable impurities are such that oxygen is 0.25 weight % or less, carbon is 0.15 weight % or less, and nitrogen is 0.03 weight % or less. Particularly preferable contents of inevitable impurities are such that oxygen is 0.05-0.25 weight %, carbon is 0.01-0.15 weight %, and nitrogen is 0.02-0.15 weight %.

[0029] Specific examples of compositions of the R-T-B sintered permanent magnets containing such inevitable  
20 impurities are as follows:

- (i) A composition comprising by weight, more than 31% and 33% or less of R, 0.6% or less of oxygen, 0.15% or less of carbon, 0.03% or less of nitrogen, and 0.3% or less of Ca. For instance, a dry molding method can provide  
25 0.25-0.6% of oxygen, 0.01-0.15% of carbon and 0.005-0.03% of nitrogen.
- (ii) A composition comprising by weight, 28-31% of R, 0.25% or less of oxygen, 0.15% or less of carbon, 0.15% or less of nitrogen, and 0.3% or less of Ca. For instance, a wet molding method can provide 0.05-0.25% of oxygen, 0.01-0.15% of carbon and 0.02-0.15% of nitrogen.

## 30 (B) Crystal structure

[0030] The crystal structure of the R-T-B sintered permanent magnet of the preset invention comprises  $R_2T_{14}B$ -type, main-phase crystal grain particles and a crystal grain boundary phase, and the  $R_2T_{14}B$ -type, main-phase crystal grain particles comprises at least (i) first  $R_2T_{14}B$ -type, main-phase crystal grain particles having a higher concentration of heavy rare earth elements than that of the crystal grain boundary phase, and (ii) second  $R_2T_{14}B$ -type, main-phase  
35 crystal grain particles having a lower concentration of heavy rare earth elements than that of the crystal grain boundary phase. The above  $R_2T_{14}B$ -type, main-phase crystal grain particles may further contain (iii) third  $R_2T_{14}B$ -type, main-phase crystal grain particles having substantially the same concentration of heavy rare earth elements as that of the crystal grain boundary phase. The concentration of heavy rare earth elements in the  $R_2T_{14}B$ -type, main-phase crystal grain particles is measured substantially in their core portions, namely substantially in their center portions. A core portion of an  $R_2T_{14}B$ -type, main-phase crystal grain particle is defined as a region of the  $R_2T_{14}B$ -type, main-phase crystal grain particle away from its crystal grain boundary by 1.0  $\mu m$  or more. The heavy rare earth element is preferably Dy, though it may be Tb and/or Ho, or their mixtures with Dy.

[0031] When the number of each type of  $R_2T_{14}B$ -type, main-phase crystal grain particles is expressed by percentage per the total number (100%) of the  $R_2T_{14}B$ -type, main-phase crystal grain particles in a cross section photograph  
45 of the crystal structure, it is preferable that the number of the first  $R_2T_{14}B$ -type, main-phase crystal grain particles is 1-35%, the number of the second  $R_2T_{14}B$ -type, main-phase crystal grain particles is 3-55%, and the number of the third  $R_2T_{14}B$ -type, main-phase crystal grain particles is 96-10%. When any of the first to third  $R_2T_{14}B$ -type, main-phase crystal grain particles are outside the above percentage range of number, it is not easy to provide the R-T-B sintered permanent magnet with high coercivity iHc, residual magnetic flux density Br and maximum energy product  $(BH)_{max}$ .  
50 More preferably, the number of the first  $R_2T_{14}B$ -type, main-phase crystal grain particles is 3-30%, the number of the second  $R_2T_{14}B$ -type, main-phase crystal grain particles is 10-45%, and the number of the third  $R_2T_{14}B$ -type, main-phase crystal grain particles is 87-25%.

## 55 [2] Production method

[0032] To produce the R-T-B sintered permanent magnet of the present invention having the above crystal structure, a so-called blend method is used, in which two types or more of R-T-B alloy powder having different concentrations of heavy rare earth elements such as Dy are mixed. In this case, the total amount of the rare earth elements does not

Table 1

Alloy	Composition (weight %)							
	Nd	Dy	B	Nb	Al	Co	Cu	Fe
A	29.0	1.0	1.0	0.2	0.1	1.0	0.1	Bal.
B	15.0	15.0	1.0	0.2	0.1	1.0	0.1	Bal.

[0040] The above mixed coarse powder was pulverized by a jet mill in a nitrogen gas atmosphere having an oxygen concentration of 10 ppm or less by volume to provide fine powder having an average diameter of 4.0  $\mu\text{m}$ . The fine powder was directly recovered in a mineral oil ("Idemitsu Super-Sol PA-30," available from Idemitsu Kosan CO., LTD.) in a nitrogen gas atmosphere without contact with the air. The resultant fine powder slurry was subjected to a wet compression molding under the conditions of a magnetic field intensity of 10 kOe and compression pressure of 1.0 ton/cm<sup>2</sup>. The resultant green body was subjected to oil removal at 200°C in a vacuum of  $5 \times 10^{-1}$  Torr for 1 hour, sintered at each temperature between 1050°C and 1100°C at about  $3 \times 10^{-5}$  Torr for 2 hours, and then cooled to room temperature.

[0041] Each of the resultant sintered bodies was heat-treated twice at 900°C for 2 hours and at 500°C for 1 hour in an inert gas atmosphere, and then cooled to room temperature to provide an R-T-B sintered permanent magnet. As a result of measuring their magnetic properties at 20°C, the results shown in Fig. 1 were obtained. As is clear from Fig. 1, preferred magnetic properties for permanent magnets were obtained at sintering temperatures of 1070-1110°C. Particularly the sintering temperature of 1090°C provided Br of 13.8 kG, iHc of 18 kOe, and (BH)<sub>max</sub> of 45.9 MGOe. Also, the sintering temperature of 1100°C provided Br of 13.8 kG, iHc of 17.9 kOe, and (BH)<sub>max</sub> of 45.7 MGOe, Br and (BH)<sub>max</sub> being high.

[0042] Analysis of a typical sintered magnet among the above sintered magnets with respect to composition indicated that the main components were 27.3 weight % of Nd, 2.7 weight % of Dy, 1.0 weight % of B, 0.2 weight % of Nb, 0.1 weight % of Al, 1.0 weight % of Co, and 0.1 weight % of Cu, the balance being substantially Fe, and that the impurities were 0.17 weight % of O, 0.05 weight % of N, and 0.07 weight % of C.

[0043] A typical sintered magnet among the above sintered magnets was observed with respect to a cross section structure in the same manner as in EXAMPLE 7 below, to determine the concentration of a heavy rare earth element (Dy) not only in main-phase crystal grain particles (R<sub>2</sub>T<sub>14</sub>B) substantially in center portions but also in a crystal grain boundary phase. As a result, it was found that the R<sub>2</sub>T<sub>14</sub>B-type, main-phase crystal grain particles were constituted by first R<sub>2</sub>T<sub>14</sub>B-type, main-phase crystal grain particles having a higher concentration of a heavy rare earth element (Dy) than that of the crystal grain boundary phase, second R<sub>2</sub>T<sub>14</sub>B-type, main-phase crystal grain particles having a lower concentration of a heavy rare earth element (Dy) than that of the crystal grain boundary phase, and third R<sub>2</sub>T<sub>14</sub>B-type, main-phase crystal grain particles having substantially the same concentration of a heavy rare earth element (Dy) as that of the crystal grain boundary phase.

#### COMPARATIVE EXAMPLE 1

[0044] Coarse pulverization was carried out in the same manner as in EXAMPLE 1 except for using a cast alloy C having a main component composition shown in Table 2. Analysis of this coarse powder with respect to composition indicated that the main components were 27.3 weight % of Nd, 2.7 weight % of Dy, 1.0 weight % of B, 0.2 weight % of Nb, 0.1 weight % of Al, 1.0 weight % of Co, and 0.1 weight % of Cu, the balance being substantially Fe, and that the impurities were 0.13 weight % of O, 0.008 weight % of N, and 0.02 weight % of C.

Table 2

Alloy	Composition (weight %)							
	Nd	Dy	B	Nb	Al	Co	Cu	Fe
C	27.3	2.7	1.0	0.2	0.1	1.0	0.1	Bal.

[0045] This coarse powder was subjected to fine pulverization to an average diameter of 4.1  $\mu\text{m}$ , forming into a slurry, molding in a magnetic field, oil removal, sintering and heat treatment in the same manner as in EXAMPLE 1, thereby providing a sintered permanent magnet of Comparative Example by a single method. Analysis of this sintered permanent magnet with respect to composition indicated that the main components were 27.3 weight % of Nd, 2.7

indicated that the main components were 22.4 weight % of Nd, 8.9 weight % of Pr, 1.2 weight % of Dy, 1.0 weight % of B, 0.1 weight % of Al, and 0.15 weight % of Ga, the balance being substantially Fe, and that the impurities were 0.14 weight % of O, 0.01 weight % of N, and 0.02 weight % of C.

Table 4

Alloy	Composition (weight %)						
	Nd	Pr	Dy	B	Al	Ga	Fe
F	22.4	8.9	1.2	1.0	0.1	0.15	Bal.

[0053] This coarse powder was subjected to fine pulverization to an average diameter of 4.0  $\mu\text{m}$ , molding in a magnetic field, sintering and heat treatment in the same manner as in EXAMPLE 2, thereby providing a sintered permanent magnet of Comparative Example by a single method. Analysis of this sintered permanent magnet with respect to composition indicated that the main components were 22.4 weight % of Nd, 8.9 weight % of Pr, 1.2 weight % of Dy, 1.0 weight % of B, 0.1 weight % of Al, and 0.15 weight % of Ga, the balance being substantially Fe, and that the impurities were 0.43 weight % of O, 0.03 weight % of N, and 0.06 weight % of C.

[0054] The measurement results of magnetic properties at 20°C are shown in Fig. 2. It is clear from Fig. 2 that through this sintered permanent magnet had slightly higher  $iH_c$  than that in EXAMPLE 2, it had as low Br as 12.9 kG or less and as low  $(BH)_{\text{max}}$  as 40.1 MGOe or less. Also, main-phase crystal grain particles having a higher concentration of a heavy rare earth element (Dy) than that of the crystal grain boundary phase were not observed in a cross section structure of the sintered magnet of this Comparative Example.

### EXAMPLE 3

[0055] Coarse pulverization was carried out in the same manner as in EXAMPLE 1 except for using cast alloys G and H having main component compositions shown in Table 5. 81.8 kg of coarse powder of the alloy G and 18.2 kg of coarse powder of the alloy H were charged into a V-type blender to provide 100 kg of mixed coarse powder. As a result of analysis of this mixed coarse powder with respect to composition, the main components of this mixed coarse powder were 19.14 weight % of Nd, 5.34 weight % of Pr, 6.00 weight % of Dy, 0.97 weight % of B, 0.29 weight % of Nb, 0.10 weight % of Al, 2.00 weight % of Co, 0.08 weight % of Ga, and 0.10 weight % of Cu, the balance being substantially Fe, and that the impurities were 0.14 weight % of O, 0.01 weight % of N, and 0.02 weight % of C.

Table 5

Alloy	Composition (weight %)									
	Nd	Pr	Dy	B	Nb	Al	Co	Ga	Cu	Fe
G	22.29	6.21	2.00	0.97	0.35	0.10	2.00	0.08	0.10	Bal.
H	5.03	1.47	24.00	0.97	-	0.10	2.00	0.08	0.10	Bal.

[0056] This mixed coarse powder was subjected to fine pulverization to an average diameter of 4.2  $\mu\text{m}$ , forming to slurry and compression-molding in a magnetic field in the same manner as in EXAMPLE 1. The resultant green body was subjected to oil removal at 200°C in a vacuum of about  $5 \times 10^{-1}$  Torr for 1 hour, sintered at each temperature between 1060°C and 1130°C at about  $2 \times 10^{-5}$  Torr for 2 hours, and then cooled to room temperature.

[0057] Each of the resultant sintered bodies was heat-treated twice at 900°C for 2 hours and at 500°C for 1 hour in an inert gas atmosphere, and then cooled to room temperature to provide an R-T-B sintered permanent magnet. As a result of measuring their magnetic properties at 20°C, the results shown in Fig. 3 were obtained. As is clear from Fig. 3, preferred magnetic properties for permanent magnets were obtained at sintering temperatures of 1070-1120°C. Particularly the sintering temperature of 1100°C provided Br of 12.7 kG,  $iH_c$  of 25.5 kOe, and  $(BH)_{\text{max}}$  of 38.8 MGOe. Also, the sintering temperature of 1110°C provided Br of 12.7 kG,  $iH_c$  of 25.3 kOe, and  $(BH)_{\text{max}}$  of 38.6 MGOe, Br and  $(BH)_{\text{max}}$  being high.

[0058] Analysis of a typical sintered magnet among the above sintered magnets with respect to composition indicated that the main components were 19.14 weight % of Nd, 5.34 weight % of Pr, 6.00 weight % of Dy, 0.97 weight % of B, 0.29 weight % of Nb, 0.10 weight % of Al, 2.00 weight % of Co, 0.08 weight % of Ga, and 0.10 weight % of Cu, the balance being substantially Fe, and that the impurities were 0.16 weight % of O, 0.05 weight % of N, and 0.07 weight



Table 7 (continued)

Alloy	Composition (weight %)									
	Nd	Pr	Dy	B	Nb	Al	Co	Ga	Cu	Fe
K	5.03	1.47	24.00	0.97	-	0.10	2.00	0.08	0.10	Bal.

[0064] This coarse powder was subjected to fine pulverization to an average diameter of 4.1  $\mu\text{m}$ , forming to slurry and molding in a magnetic field in the same manner as in EXAMPLE 1. The resultant green body was subjected to oil removal at 200°C in a vacuum of about  $5 \times 10^{-1}$  Torr for 1 hour, sintered at each temperature between 1060°C and 1130°C at about  $2 \times 10^{-5}$  Torr for 2 hours, and then cooled to room temperature.

[0065] Each of the resultant sintered bodies was heat-treated twice at 900°C for 2 hours and at 500°C for 1 hour in an inert gas atmosphere, and then cooled to room temperature to provide a sintered permanent magnet of Comparative Example by a blend method. As a result of measuring their magnetic properties at 20°C, the results shown in Fig. 3 were obtained. As is clear from Fig. 3, the sintering temperature of 1100°C provided Br of 12.1 kG, iHc of 25.4 kOe, and (BH)<sub>max</sub> of 35.1 MGOe. Also, the sintering temperature of 1110°C provided Br of 12.1 kG, iHc of 25.2 kOe, and (BH)<sub>max</sub> of 35.0 MGOe, Br and (BH)<sub>max</sub> being low.

[0066] Analysis of the sintered magnet of this Comparative Example with respect to composition indicated that the main components were 19.14 weight % of Nd, 5.34 weight % of Pr, 6.00 weight % of Dy, 0.97 weight % of B, 0.65 weight % of Nb, 0.10 weight % of Al, 2.00 weight % of Co, 0.08 weight % of Ga, and 0.10 weight % of Cu, the balance being substantially Fe, and that the impurities were 0.17 weight % of O, 0.06 weight % of N, and 0.06 weight % of C. Why the sintered magnet of this Comparative Example has low Br and (BH)<sub>max</sub> appears to be due to the fact that normal growth of main-phase crystal grain particles during sintering is hindered by as high a Nb content as 0.65 weight %.

#### EXAMPLE 4

[0067] Coarse pulverization was carried out in the same manner as in EXAMPLE 1 except for using cast alloys L and M having main component compositions shown in Table 8. 90.0 kg of coarse powder of the alloy L and 10.0 kg of coarse powder of the alloy M were charged into a V-type blender to provide 100 kg of mixed coarse powder. As a result of analysis of this mixed coarse powder with respect to composition, the main components of this mixed coarse powder were 22.83 weight % of Nd, 6.37 weight % of Pr, 1.30 weight % of Dy, 1.05 weight % of B, 0.13 weight % of Mo, and 0.10 weight % of Al, the balance being substantially Fe, and that the impurities were 0.15 weight % of O, 0.01 weight % of N, and 0.02 weight % of C.

Table 8

Alloy	Composition (weight %)						
	Nd	Pr	Dy	B	Mo	Al	Fe
L	23.85	6.65	-	1.05	0.15	0.10	Bal.
M	13.68	3.82	13.00	1.05	-	0.10	Bal.

[0068] This mixed coarse powder was subjected to fine pulverization to an average diameter of 4.0  $\mu\text{m}$ , forming to slurry and molding in a magnetic field in the same manner as in EXAMPLE 1. The resultant green body was subjected to oil removal at 200°C in a vacuum of about  $5 \times 10^{-1}$  Torr for 1 hour, sintered at each temperature between 1050°C and 1100°C in vacuum of about  $2 \times 10^{-5}$  Torr for 2 hours, and then cooled to room temperature.

[0069] Each of the resultant sintered bodies was heat-treated twice at 900°C for 2 hours and at 550°C for 1 hour in an inert gas atmosphere, and then cooled to room temperature to provide an R-T-B sintered permanent magnet. As a result of measuring their magnetic properties at 20°C, it was found that preferred magnetic properties for permanent magnets were obtained at sintering temperatures of 1060-1090°C. Particularly the sintering temperature of 1070°C provided Br of 13.9 kG, iHc of 15.5 kOe, and (BH)<sub>max</sub> of 46.5 MGOe. Also, the sintering temperature of 1080°C provided Br of 14.0 kG, iHc of 15.3 kOe, and (BH)<sub>max</sub> of 47.2 MGOe, Br and (BH)<sub>max</sub> being high.

[0070] Analysis of a typical sintered magnet among the above sintered magnets with respect to composition indicated that the main components were 22.83 weight % of Nd, 6.37 weight % of Pr, 1.30 weight % of Dy, 1.05 weight % of B, 0.13 weight % of Mo, and 0.10 weight % of Al, the balance being substantially Fe, and that the impurities were 0.18 weight % of O, 0.06 weight % of N, and 0.08 weight % of C.

[0071] With respect to cross section structures of the above permanent magnets produced at sintering tempera-

mixed coarse powder were 20.6 weight % of Nd, 8.8 weight % of Pr, 2.6 weight % of Dy, 1.06 weight % of B, 0.18 weight % of W, 0.05 weight % of Al, and 0.17 weight % of Ga, the balance being substantially Fe, and that the impurities were 0.15 weight % of O, 0.01 weight % of N, and 0.01 weight % of C.

Table 10

Alloy	Composition (weight %)							
	Nd	Pr	Dy	B	W	Al	Ga	Fe
P	21.70	9.30	1.00	1.06	0.20	0.05	0.17	Bal.
Q	10.50	4.50	17.00	1.06	-	0.05	0.17	Bal.

[0077] The above mixed coarse powder was pulverized by a jet mill in a nitrogen gas atmosphere having an oxygen concentration of 500 ppm or less by volume to provide fine powder having an average diameter of 4.2  $\mu\text{m}$ . The fine powder was subjected to a dry compression molding under the conditions of a magnetic field intensity of 10 kOe and compression pressure of 1.5 ton/cm<sup>2</sup>. The resultant green body was sintered at each temperature between 1040°C and 1100°C at about  $3 \times 10^{-5}$  Torr for 2 hours, and then cooled to room temperature.

[0078] Each of the resultant sintered bodies was heat-treated twice at 900°C for 3 hours and at 550°C for 1 hour in an inert gas atmosphere, and then cooled to room temperature to provide an R-T-B sintered permanent magnet. As a result of measuring their magnetic properties at 20°C, it was found that preferred magnetic properties for permanent magnets were obtained at sintering temperatures of 1050-1090°C. Particularly the sintering temperature of 1070°C provided Br of 13.2 kG, iHc of 19.5 kOe, and (BH)<sub>max</sub> of 41.8 MGOe. Also, the sintering temperature of 1080°C provided Br of 13.2 kG, iHc of 19.3 kOe, and (BH)<sub>max</sub> of 41.7 MGOe, Br and (BH)<sub>max</sub> being high.

[0079] Analysis of a typical permanent magnet among the above permanent magnets with respect to composition indicated that its main components were 20.6 weight % of Nd, 8.8 weight % of Pr, 2.6 weight % of Dy, 1.06 weight % of B, 0.18 weight % of W, 0.05 weight % of Al, and 0.17 weight % of Ga, the balance being substantially Fe, and that the impurities were 0.50 weight % of O, 0.02 weight % of N, and 0.06 weight % of C.

[0080] With respect to cross section structures of the above permanent magnets produced at sintering temperatures of 1070°C and 1080°C, respectively, the concentration of a heavy rare earth element (Dy) was measured in main-phase crystal grain particles (R<sub>2</sub>T<sub>14</sub>B) substantially in center portions and a crystal grain boundary phase in the same manner as in EXAMPLE 7 below. As a result, it was found that the R<sub>2</sub>T<sub>14</sub>B-type, main-phase crystal grain particles were constituted by first R<sub>2</sub>T<sub>14</sub>B-type, main-phase crystal grain particles having a higher concentration of a heavy rare earth element (Dy) than that of the crystal grain boundary phase, second R<sub>2</sub>T<sub>14</sub>B-type, main-phase crystal grain particles having a lower concentration of a heavy rare earth element (Dy) than that of the crystal grain boundary phase, and third R<sub>2</sub>T<sub>14</sub>B-type, main-phase crystal grain particles having substantially the same concentration of a heavy rare earth element (Dy) as that of the crystal grain boundary phase.

#### EXAMPLE 7

[0081] Coarse pulverization was carried out in the same manner as in EXAMPLE 1 except for using cast alloys R and S having main component compositions shown in Table 11. 90.0 kg of coarse powder of the alloy R and 10.0 kg of coarse powder of the alloy S were charged into a V-type blender to provide 100 kg of mixed coarse powder. As a result of analysis of this mixed coarse powder with respect to composition, it was found that the main components of this mixed coarse powder were 21.38 weight % of Nd, 7.12 weight % of Pr, 1.50 weight % of Dy, 1.03 weight % of B, 0.08 weight % of Al, 2.00 weight % of Co, 0.08 weight % of Ga, and 0.1 weight % of Cu, the balance being substantially Fe, and that the impurities were 0.14 weight % of O, 0.02 weight % of N, and 0.02 weight % of C.

Table 11

Alloy	Composition (weight %)								
	Nd	Pr	Dy	B	Al	Co	Ga	Cu	Fe
R	22.50	7.50	-	1.03	0.08	2.00	0.08	0.10	Bal.
S	11.25	3.75	15.00	1.03	0.08	2.00	0.08	0.10	Bal.

[0082] The above mixed coarse powder was pulverized by a jet mill in a nitrogen gas atmosphere having an oxygen

taken in a lot of visual fields should be averaged. Thus, the R-T-B sintered permanent magnet of the present invention has a characteristic concentration distribution of Dy in the main-phase crystal grain particles and the crystal grain boundary phase.

[0090] With respect to a typical permanent magnet among the above permanent magnets, a particle size distribution of the main-phase crystal grain particles is shown in Fig. 5. In Fig. 5, the axis of abscissas represents a particle size range of main-phase crystal grain particles. For instance, "9-10  $\mu\text{m}$ " means that the particle size range of main-phase crystal grain particles is 9  $\mu\text{m}$  or more and less than 10  $\mu\text{m}$ . The particle size of main-phase crystal grain particles was determined by taking a photomicrograph (magnification: 1000) of an arbitrary cross section of the permanent magnet by an optical microscope (UFX-II, available from Nikon), and image-treating this photomicrograph by an image treatment software (Image Pro. Plus (DOS/V), available from Planetron). Assuming that each main phase crystal grain particle has a circular cross section having an area  $S_i$  measured by image treatment, a particle size  $d_i$  of each main phase crystal grain particle is defined as  $(4 \times S_i \div \pi)^{1/2}$ . The axis of ordinates represents a distribution ratio (%), a ratio of the number  $T_N$  of main-phase crystal grain particles in each particle size range to the total number  $T$  of main-phase crystal grain particles in a visual field measured:  $[(T_N/T) \times 100\%]$ .

[0091] As is clear from Fig. 5, in the permanent magnet of the present invention, the distribution ratio of main-phase crystal grain particles was 0% in a particle size range of less than 2  $\mu\text{m}$ , and 5.8% in a particle size range of 16  $\mu\text{m}$  or more. Further investigation has revealed that when the distribution ratio of main-phase crystal grain particles is less than 5% in a particle size range of less than 2  $\mu\text{m}$  and 10% or less in a particle size range of 16  $\mu\text{m}$  or more, preferred magnetic properties as permanent magnets can be obtained. Further, it is preferable that the distribution ratio of main-phase crystal grain particles is 3% or less in a particle size range of less than 2  $\mu\text{m}$  and 8% or less in a particle size range of 16  $\mu\text{m}$  or more. Particularly preferable is that the distribution ratio of main-phase crystal grain particles is 0% in a particle size range of less than 2  $\mu\text{m}$  and 6% or less in a particle size range of 16  $\mu\text{m}$  or more. Incidentally, the above particle size distribution of main-phase crystal grain particles can be achieved even in the case of the Nb content of 0.01-0.6 weight %.

#### COMPARATIVE EXAMPLE 5

[0092] Coarse pulverization was carried out in the same manner as in EXAMPLE 7 except for using a cast alloy T having a main component composition shown in Table 12. As a result of analysis of this coarse powder with respect to composition, it was found that the main components of this coarse powder were 21.38 weight % of Nd, 7.12 weight % of Pr, 1.50 weight % of Dy, 1.03 weight % of B, 0.70 weight % of Nb, 0.08 weight % of Al, 2.00 weight % of Co, 0.08 weight % of Ga, and 0.1 weight % of Cu, the balance being substantially Fe, and that the impurities were 0.15 weight % of O, 0.01 weight % of N, and 0.02 weight % of C.

Table 12

Alloy	Composition (weight %)									
	Nd	Pr	Dy	B	Nb	Al	Co	Ga	Cu	Fe
T	21.38	7.12	1.50	1.03	0.70	0.08	2.00	0.08	0.10	Bal.

[0093] This coarse powder was subjected to fine pulverization to an average diameter of 4.1  $\mu\text{m}$ , forming into a slurry, molding in a magnetic field, oil removal, sintering and heat treatment in the same manner as in EXAMPLE 7, thereby providing a sintered permanent magnet of Comparative Example by a single method. Analysis of this sintered permanent magnet with respect to composition indicated that the main components were 21.38 weight % of Nd, 7.12 weight % of Pr, 1.50 weight % of Dy, 1.03 weight % of B, 0.70 weight % of Nb, 0.08 weight % of Al, 2.00 weight % of Co, 0.08 weight % of Ga, and 0.1 weight % of Cu, the balance being substantially Fe, and that the impurities were 0.17 weight % of O, 0.05 weight % of N, and 0.07 weight % of C.

[0094] As a result of measuring their magnetic properties at 20°C, it was found that though this sintered permanent magnet had as high  $iH_c$  as about 16 kOe, it had  $B_r$  of 13.5 kG or less and  $(BH)_{\text{max}}$  of 44.0 MGOe or less, smaller than those in EXAMPLE 7.

[0095] Fig. 6 schematically shows the cross section structure of this sintered magnet. In the crystal structure, 3 denotes voids, and other numerals denote the same parts as in Fig. 4(a). It is clear from Fig. 6 that there were two patterns of concentration distributions of Dy, one that was substantially uniform from the crystal grain boundary phase to center portions of the main-phase crystal grain particles, and the other in which the concentration distribution of Dy was higher in the crystal grain boundary phase and lower substantially in center portions of the main-phase crystal grain particles. The number of the main-phase crystal grain particles having substantially the same Dy concentration distri-



3. The R-T-B sintered permanent magnet according to claim 1 having a composition comprising 28-33 weight % of R, 0.5-2 weight % of B, 0.01-0.6 weight % of  $M_1$ , and 0.01-0.3 weight % of  $M_2$ , the balance being substantially T and inevitable impurities, wherein  $M_1$  is at least one element selected from the group consisting of Nb, Mo, W, V, Ta, Cr, Ti, Zr and Hf, and  $M_2$  is at least one element selected from the group consisting of Al, Ga and Cu.

4. The R-T-B sintered permanent magnet according to any one of claims 1-3, wherein the amount of said R is more than 31 % and 33 % or less by weight.

5. The R-T-B sintered permanent magnet according to any one of claims 1-4, comprising as inevitable impurities 0.6 weight % or less of oxygen, 0.15 weight % or less of carbon, 0.03 weight % or less of nitrogen and 0.3 weight % or less of Ca.

6. The rare earth magnet material according to any one of claims 1-4, comprising as inevitable impurities 0.25 weight % or less of oxygen, 0.15 weight % or less of carbon, 0.15 weight % or less of nitrogen and 0.3 weight % or less of Ca.

Fig. 2

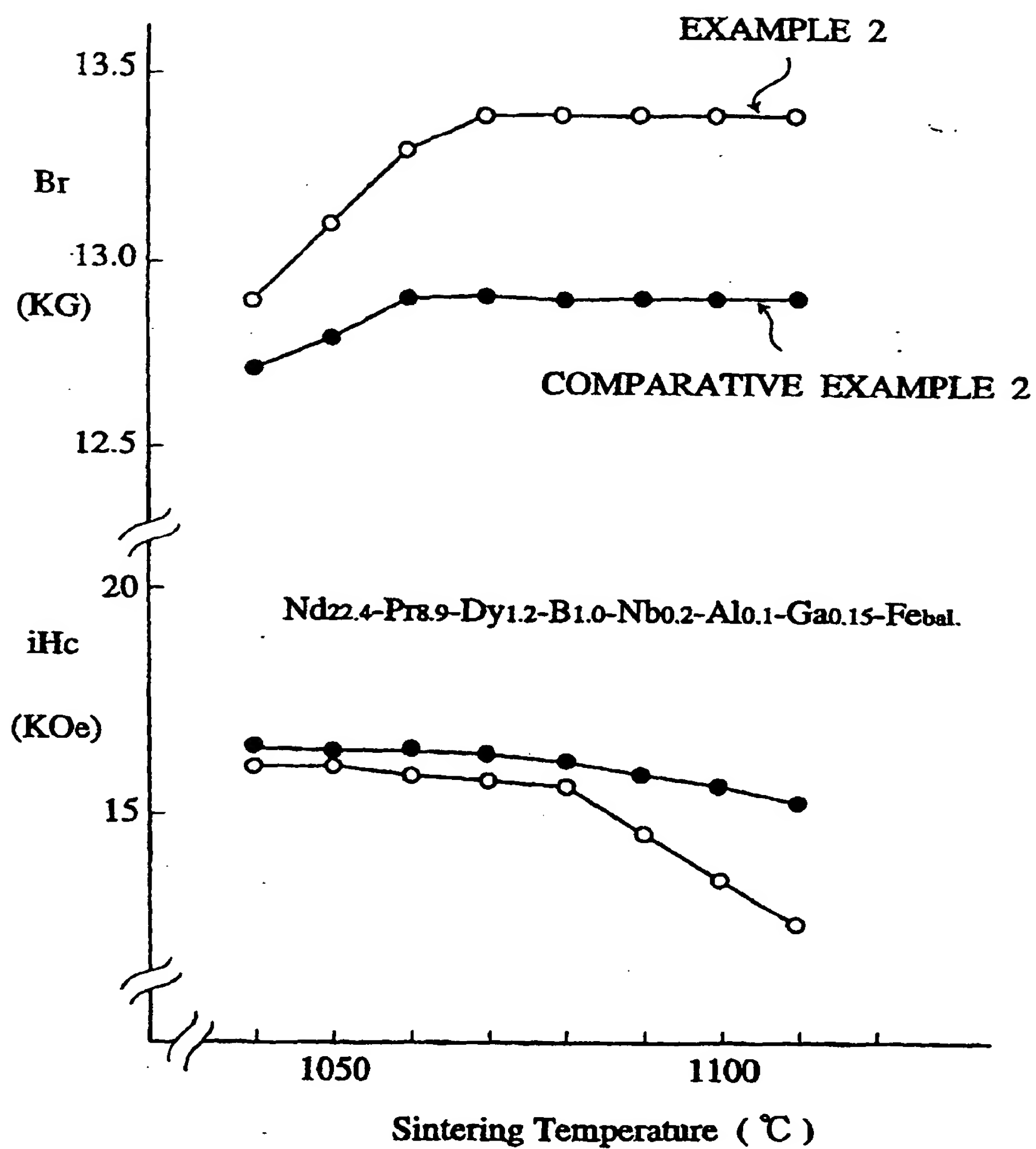


Fig. 4(a)

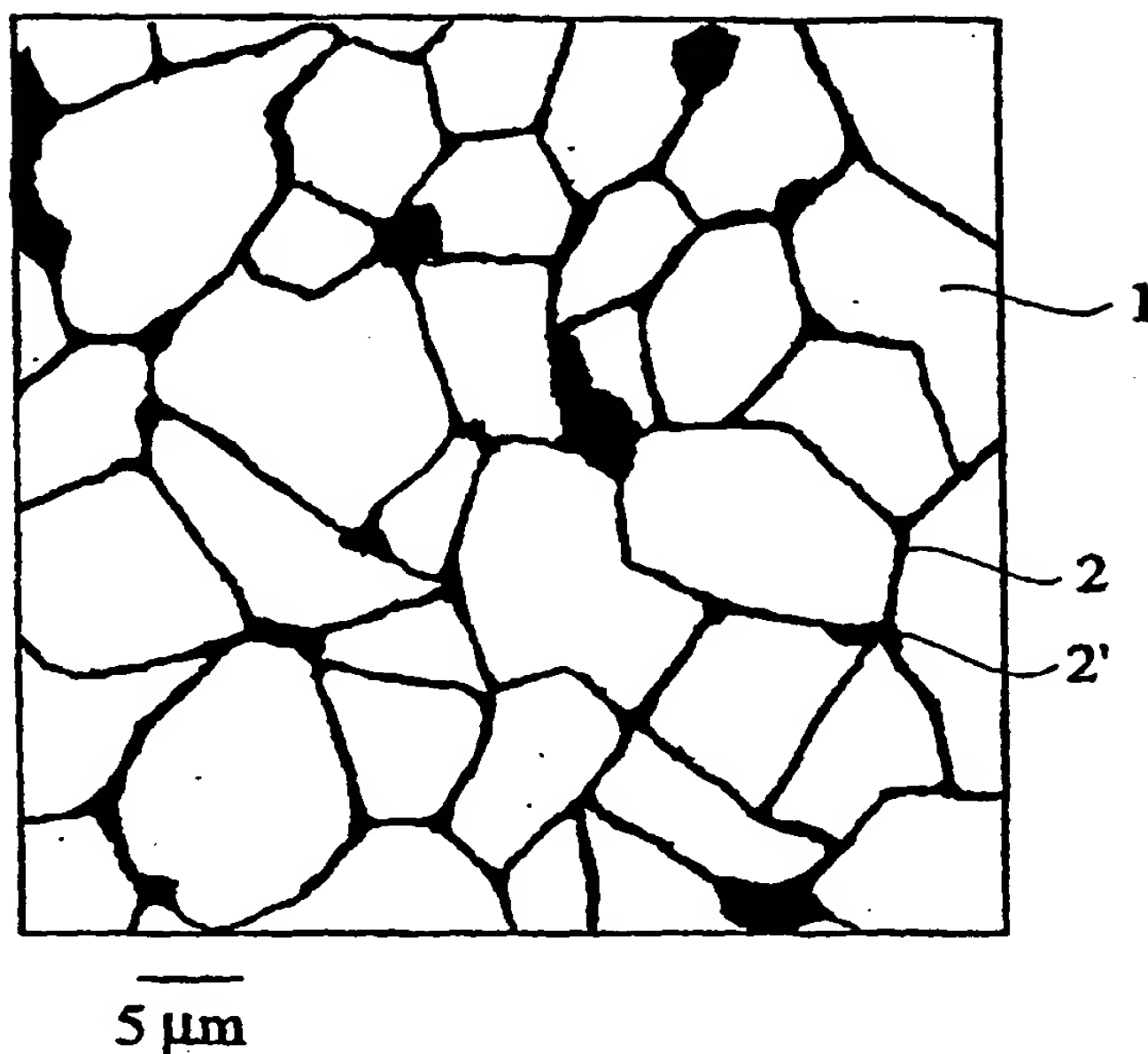


Fig. 4(b)

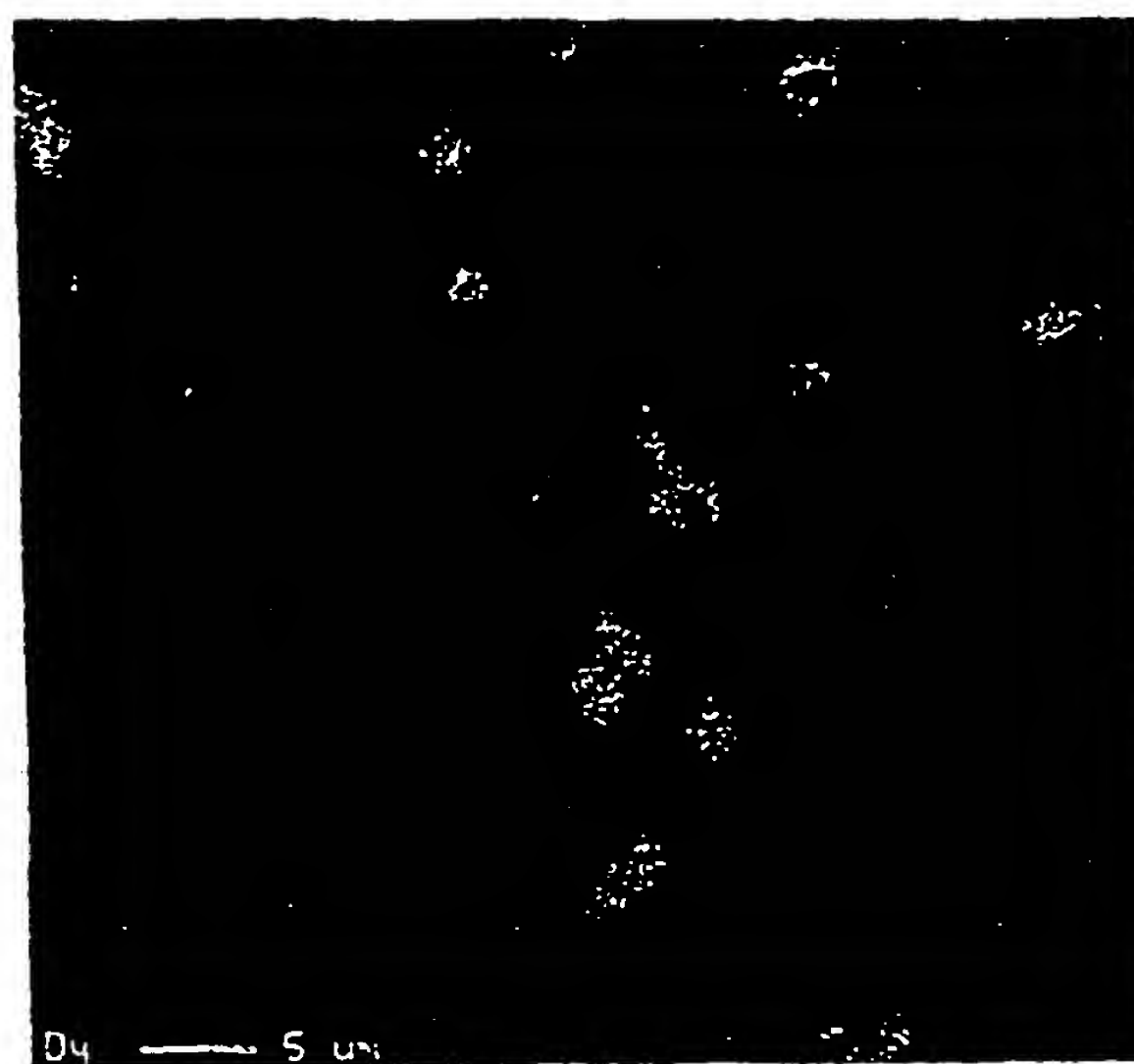


Fig. 5

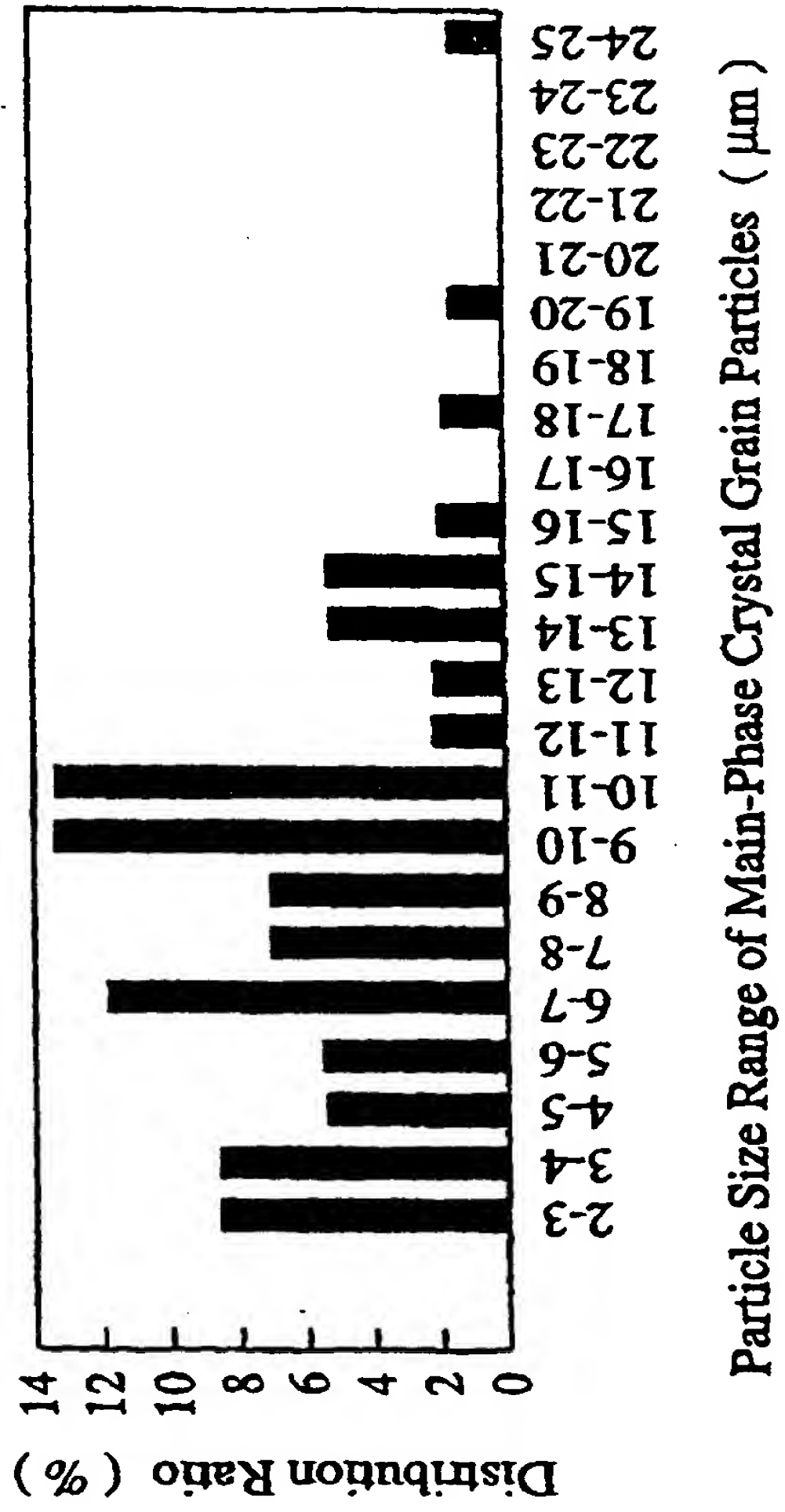
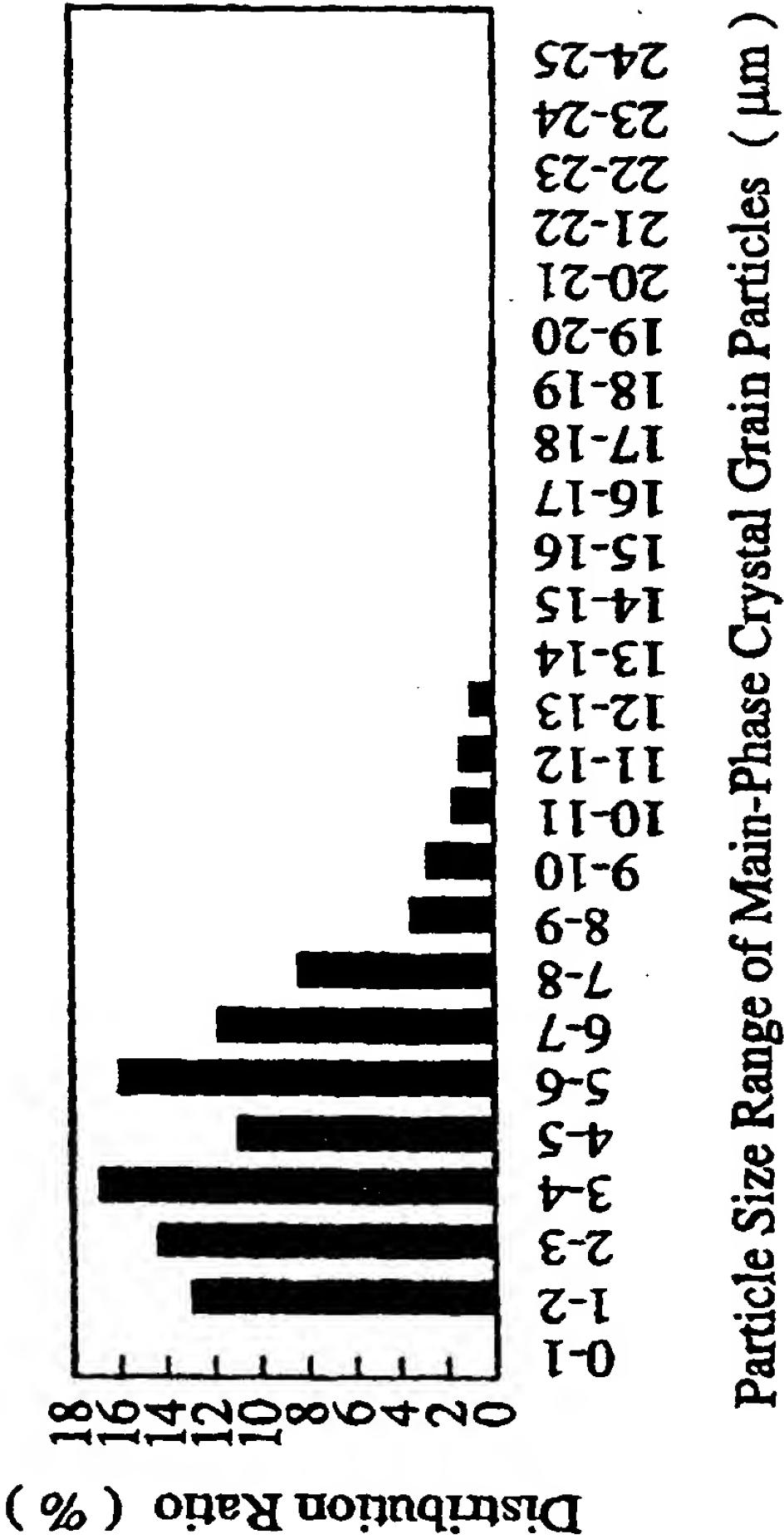
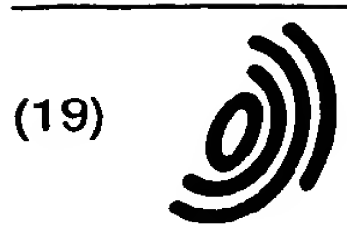




Fig. 7





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(54) **R-T-B sintered permanent magnet**

(57) An R-T-B sintered permanent magnet having a composition including 28-33 weight % of R, and 0.5-2 weight % of B, the balance being substantially T and inevitable impurities, wherein R is at least one rare earth element including Y, at least one heavy rare earth element selected from the group consisting of Dy, Tb and Ho being indispensable, and T is Fe or Fe and Co, the permanent magnet having a crystal structure comprising first R<sub>2</sub>T<sub>14</sub>B-type, main-phase crystal grain particles having a higher heavy rare earth element concentration than that of a crystal grain boundary phase, and second R<sub>2</sub>T<sub>14</sub>B-type, main-phase crystal grain particles having a lower heavy rare earth element concentration than that of the crystal grain boundary phase.

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